Possible Realization of Odd-Frequency Pairing in Heavy Fermion Compounds

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Using Majorana fermions to represent spins we reexamine the Kondo lattice model for heavy fermions. The simplest decoupling procedure provides a realization of odd-frequency superconductivity, with resonant pairing and surfaces of gap zeros. Spin and charge coherence factors vanish linearly with the energy on the Fermi surface, predicting a linear specific heat, but a T^3 NMR relaxation rate. Possible application to heavy fermions is suggested.

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Though a decade and a half has passed since the discovery of the heavy fermion metals and superconductors [1,2], many experimental anomalies remain. While the basic theoretical picture of resonantly scattered conduction electrons forming a highly renormalized f band is not in question, certain experimental features fit awkwardly into the standard model [3-7]. The underlying nature of the interactions [8], the nature of the pairing [9], and the excitation spectrum of heavy fermion insulators [10] are three areas of continuing uncertainty.

Conventional approaches to heavy fermion physics represent the f moments as fermions by enforcing a "Gutzwiller constraint" of unit occupancy $n_f=1$ at each site. This requires a projection of the physical Hilbert space of the local moments from the larger Hilbert space of pseudo fermions: a task that is difficult to do exactly, and usually only treated on the average. In this Letter we examine an approach to a simple Kondo lattice model for heavy fermions that avoids these difficulties.

Various new features are predicted that differ qualitatively from the standard model of heavy fermion behavior; most notably a development of strong correlations between the spin and pair degrees of freedom, forming a ground state where the conduction electrons experience frequency dependent or "resonant" triplet pairing. The pairing fields actually diverge at low frequencies as the inverse frequency, providing a first stable realization of the phenomenon of odd-frequency pairing originally considered by Berezinskii [11,12]. For a wide range of conditions, including the presence of spin-orbit coupling, this theory predicts surfaces of gapless excitations, and a linear specific heat that survives in the superconducting state. Unlike a conventional superconductor, the charge and spin coherence factors vanish on the pseudo Fermi surface, giving rise to a T^3 NMR relaxation. In this scenario, the linear specific heat anomalies often observed in heavy fermion superconductors [13] might be interpreted as intrinsic.

A key feature of our approach is the use of a special anticommuting representation of spin-$\frac{1}{2}$ operators to describe the magnetic excitations within the low-lying crystal field doublets of the heavy fermion ions [14]. Recall that for individual $S=\frac{1}{2}$ objects, the Pauli matrices are anticommuting variables $\{\sigma_a,\sigma_b\}=2\delta_{ab}$ and consequently can be treated as real or Majorana $(\boldsymbol{\sigma}^\dagger=\boldsymbol{\sigma})$ Fermi fields. Their Fermi statistics alone guarantee that the spin operator $\mathbf{S}=\frac{1}{4}i\boldsymbol{\sigma}\times\boldsymbol{\sigma}$ satisfies both the SU(2) algebra $[S^a,S^b]=i\epsilon_{abc}S^c$ *and* the condition $\mathbf{S}^2=\frac{3}{4}$. This feature can be generalized to many sites, introducing a set of three component anticommuting real vectors $\boldsymbol{\eta}_i$ at each site i,

$$\{\eta_i^a,\eta_j^b\}=\delta_{ij}\delta^{ab}\ (\eta_j^a=\eta_j^{\dagger a})\ (a,b=1,2,3) \tag{1}$$

from which the spin operator at each site is constructed

$$\mathbf{S}_j=\frac{1}{2}i\boldsymbol{\eta}_j\times\boldsymbol{\eta}_j\,. \tag{2}$$

This "Majorana" representation of spin-$\frac{1}{2}$ operators has a long history [15] in particle physics. Loosely speaking, the Majorana fermions may be considered to be lattice generalizations of *anticommuting* Pauli operators $\boldsymbol{\eta}_j\equiv(1/\sqrt{2})\boldsymbol{\sigma}_j$. There is no constraint associated with this representation, for the spin algebra *and* the condition $S=\frac{1}{2}$ are satisfied at each site, between all states of the Fock space [16,17]. In momentum space, the Bloch waves, $\boldsymbol{\eta}_{\mathbf{k}}=\sum_j\eta_j e^{-i\mathbf{k}\cdot\mathbf{R}_j}$ behave as conventional complex fermions, but since $\eta_{\mathbf{k}}^\dagger=\eta_{-\mathbf{k}}$, the momentum lies in one half of the Brillouin zone. Finally note that since there is no constraint, the trial ground-state energy obtained from a trial Hamiltonian is a *strict variational upper bound* on the true ground-state energy.

Our basic model for a heavy fermion system is a spin-$\frac{1}{2}$ Kondo lattice model, with a single band interacting with local f moments $\mathbf{S}_j$ in each unit cell. Our simplified Hamiltonian is written

$$H=H_c+\sum_j H_{\rm int}[j]\,. \tag{3}$$

Here $H_c=\sum\epsilon_{\mathbf{k}}\psi_{\mathbf{k}}^\dagger\psi_{\mathbf{k}}$ describes the conduction band, and $\psi_{\mathbf{k}}^\dagger=(\psi_{\mathbf{k}\uparrow}^\dagger,\psi_{\mathbf{k}\downarrow}^\dagger)$ is a conduction electron spinor. The exchange interaction at each site j is written in a tight binding representation as

$$H_{\rm int}[j]=J(\psi_{j\alpha}^\dagger\boldsymbol{\sigma}_{\alpha\beta}\psi_{j\beta})\cdot\mathbf{S}_j\longrightarrow-\frac{1}{2}J\psi_j^\dagger[\boldsymbol{\sigma}_j\cdot\boldsymbol{\eta}_j]^2\psi_j\,.$$

In a real heavy fermion system, we envisage that the indices would refer to the conserved pseudospin indices of the low lying magnetic manifold. We have suppressed both the momentum dependence and anisotropy of the coupling, using $i\boldsymbol{\sigma}\cdot(\boldsymbol{\eta}\times\boldsymbol{\eta})=[\boldsymbol{\eta}\cdot\boldsymbol{\sigma}]^2-\frac{3}{2}$ to simplify the interaction.

We may now write the partition function as a path integral, $Z=\int_P e^{-\int_0^\beta \mathcal{L}(\tau)d\tau}$ where

$$\mathcal{L}(\tau)=\sum_{\mathbf{k}}\psi^\dagger_{\mathbf{k}}\partial_\tau\psi_{\mathbf{k}}+\sum_{\mathbf{k}\in\frac{1}{2}\mathrm{BZ}}\eta^\dagger_k\partial_\tau\eta_{\mathbf{k}}+H_c+\sum_j H_{\mathrm{int}}[j]\,. \tag{4}$$

Here we have factorized the interaction in terms of a fluctuating two-component spinor $V^\dagger_j=(V^*_\uparrow,V^*_\downarrow)$

$$H_{\mathrm{int}}[j]=\psi^\dagger_j(\boldsymbol{\sigma}\cdot\boldsymbol{\eta}_j)V_j+V^\dagger_j(\boldsymbol{\sigma}\cdot\boldsymbol{\eta}_j)\psi_j+2|V_j|^2/J\,. \tag{5}$$

We are particularly interested in examining static mean field solutions where

$$V_j=\frac{V}{\sqrt{2}}\begin{pmatrix} z_{j\uparrow}\\ z_{j\downarrow}\end{pmatrix},\quad z^\dagger_j z_j=1\,. \tag{6}$$

To gain insight into this mean field theory, let us integrate out the localized spin degrees of freedom, represented by the Majorana fermions. This introduces a resonant self-energy into the electron propagators, containing an isotropic component that builds the renormalized heavy fermion band and an anisotropic term, compactly represented by the effective action

$$S_c=\sum_{\{\mathbf{k},i\omega_n\}}\psi^\dagger_{\mathbf{k},\omega}[-\omega+\epsilon_{\mathbf{k}}+\Delta(\omega)]\psi_{\mathbf{k},\omega}+S_a\,, \tag{7}$$

where $\Delta(\omega)=V^2/2\omega$ determines the strength of the resonant scattering. The anisotropic term S_a is written in a tight binding basis as

$$S_a=-\sum_{\{j,i\omega_n\}}\frac{\Delta(\omega)}{2}\{\psi^\dagger_{j,\omega}[1+\mathbf{b}_j\cdot\boldsymbol{\sigma}]\psi_{j,\omega}+[\psi_{j,-\omega}[i\sigma_2\boldsymbol{\sigma}\cdot\mathbf{d}_j]\psi_{j,\omega}+\mathrm{c.c.}]\}\,. \tag{8}$$

Here the triad of orthogonal unit vectors $\hat{\mathbf{b}}=z^\dagger\boldsymbol{\sigma}z$, $\hat{\mathbf{d}}=\hat{\mathbf{x}}+i\hat{\mathbf{y}}=z^T[i\sigma_2\boldsymbol{\sigma}]z$ define the orientation of the order parameter. The quantities

$$\mathbf{B}_j(\omega)=\frac{\Delta(\omega)}{2}\hat{\mathbf{b}}_j,\quad \boldsymbol{\Delta}_j(\omega)=\frac{\Delta(\omega)}{2}\hat{\mathbf{d}}_j\,, \tag{9}$$

may be interpreted as resonant Weiss and triplet pairing fields, respectively. Unlike earlier realizations of odd-frequency triplet pairing [11,12], here the odd-frequency pairing field diverges at zero frequency, coupling spin and triplet pair degrees of freedom in one order parameter.

To simplify further discussion, we consider the case of a bipartite lattice. Here, a stable mean field solution is obtained with a *staggered* order parameter, where for example $\hat{\mathbf{b}}$ is constant, and $\hat{\mathbf{d}}=e^{i\mathbf{Q}\cdot\mathbf{R}_j}\hat{\mathbf{d}}_0$ is staggered commensurately with $\mathbf{Q}=(\pi,\pi,\pi)$. In this case the spinor $z_j=e^{i\mathbf{Q}\cdot\mathbf{R}_j/2}z_0$, where

$$z_0=\begin{pmatrix}1\\0\end{pmatrix}$$

Writing the conduction electron spinors in terms of their four real components $\chi^\lambda(\mathbf{k})$ ($\lambda=0,1,2,3$)

$$\psi_j=\frac{1}{\sqrt{2}}\{\chi^0_j+i\boldsymbol{\chi}_j\cdot\boldsymbol{\sigma}\}z_0\,, \tag{10}$$

the mean field Hamiltonian takes the simple form

$$H_{\mathrm{MF}}=\sum_{\mathbf{k}\in\frac{1}{2}\mathrm{BZ}}\{\tilde{\epsilon}_{\mathbf{k}}\chi^{\lambda\dagger}_{\mathbf{k}}\chi^\lambda_{\mathbf{k}}+iV[\boldsymbol{\eta}^\dagger_{\mathbf{k}}\cdot\boldsymbol{\chi}_{\mathbf{k}}-\mathrm{c.c.}]+\alpha_{\mathbf{k}}n_{\mathbf{k}}\}\,, \tag{11}$$

where

$$n_{\mathbf{k}}=i[(\chi^{3\dagger}_{\mathbf{k}}\chi^0_{\mathbf{k}}+\chi^{2\dagger}_{\mathbf{k}}\chi^1_{\mathbf{k}})-\mathrm{c.c.}] \tag{12}$$

is the number operator of the state $\mathbf{k}$, written in the four-component basis, and

$$\tilde{\epsilon}_{\mathbf{k}}=\tfrac{1}{2}(\epsilon_{\mathbf{k}+\mathbf{Q}/2}-\epsilon_{-\mathbf{k}+\mathbf{Q}/2})\,,\qquad \alpha_{\mathbf{k}}=\tfrac{1}{2}(\epsilon_{\mathbf{k}+\mathbf{Q}/2}+\epsilon_{-\mathbf{k}+\mathbf{Q}/2})=-\mu\,, \tag{13}$$

where the last equality holds only for a tight binding model. Let us begin by considering the special case of half filling ($\mu=0$), for in this case the Hamiltonian is diagonal in the Majorana components λ, with excitation energies

$$E_{\mathbf{k}i}=\frac{\tilde{\epsilon}_{\mathbf{k}}}{2}\pm\left[\left(\frac{\tilde{\epsilon}_{\mathbf{k}}}{2}\right)^2+V^2\right]^{1/2}\quad(i=1,3)\,,\qquad E_{\mathbf{k}0}=\tilde{\epsilon}_{\mathbf{k}}\,, \tag{14}$$

corresponding to three hybridized gapful branches and a fourth gapless Majorana mode formed from a component of the conduction band that does not mix with the local moments. With one unpaired Majorana fermion per unit cell, the corresponding Fermi surface $\tilde{\epsilon}_{\mathbf{k}}=0$ spans precisely one half of the Brillouin zone: $V_{\mathrm{FS}}/(2\pi)^3=\frac{1}{2}$. This counting argument guarantees that the gapless Fermi surface persists in the presence of particle hole asymmetry ($\mu\neq 0$) or a spin-dependent kinetic energy associated with spin-orbit coupling.

For our particular choice of z_0, the up electrons are "paired," while the "down" electrons are unpaired with a gapped excitation spectrum (Fig. 1). In a Nambu notation, their propagators are

$$G_\sigma(\omega,\mathbf{k})=\begin{cases}[(\omega-\underline{\epsilon}_{\mathbf{k}}-\Delta(\omega)(1-\underline{\tau}_1)]^{-1} & (\sigma=\uparrow),\\ [(\omega-\underline{\epsilon}_{\mathbf{k}}-2\Delta(\omega)]^{-1} & (\sigma=\downarrow)\,,\end{cases} \tag{15}$$

where $\underline{\epsilon}_{\mathbf{k}}=\tilde{\epsilon}_{\mathbf{k}}-\mu\underline{\tau}_3$. The density of states for the "up" electrons is

$$\rho_\uparrow(\omega)=\begin{cases}\frac{1}{2}\rho(1+\mu\omega/[\frac{1}{4}V^4+\mu^2\omega^2]^{1/2}) & (|\omega|<T_K),\\ \rho & (|\omega|>T_K)\,,\end{cases} \tag{16}$$

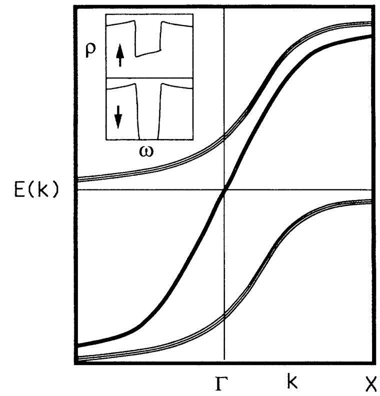

FIG. 1. Excitation spectrum of mean field theory for $\mu=0$, showing threefold degenerate gapped excitations and a gapless Majorana band. Inset: Conduction electron density of states for up (↑) and down (↓) electrons.

where $T_K=V^2/D[1-(\mu/D)^2]$ is the indirect gap associated with the excitation spectrum (14).

Unlike conventional pairing, the charge and spin coherence factors of these "Majorana" quasiparticles are strongly energy dependent. Near the Fermi surface, the gapless quasiparticle operators can be written as

$$a^\dagger_{\mathbf{k}\uparrow}=Z^{1/2}[u_{\mathbf{k}}\psi^\dagger_{\mathbf{k}\uparrow}+v_{\mathbf{k}}\psi_{-\mathbf{k}\uparrow}]+(1-Z)^{1/2}\eta^\dagger_{3\mathbf{k}}, \quad (17)$$

where $Z^{-1}=1+\mu^2/V^2$ is a quasiparticle renormalization constant and the Bogoliubov coefficients are determined by $u^2+v^2=1$,

$$u^2_{\mathbf{k}}=\frac{1}{2}\left[1+\frac{\mu\,\mathrm{sgn}(E_{\mathbf{k}})}{[\Delta(E_{\mathbf{k}})^2+\mu^2]^{1/2}}\right]. \quad (18)$$

Spin and charge coherence factors are then given by

$$\left.\begin{matrix}\langle\mathbf{k}^-|\rho_{\mathbf{q}}|\mathbf{k}^+\rangle\\ \langle\mathbf{k}^-|\sigma^z_{\mathbf{q}}|\mathbf{k}^+\rangle\end{matrix}\right\}=Z[\mu_{\mathbf{k}^-}u_{\mathbf{k}^+}-v_{\mathbf{k}^-}v_{\mathbf{k}^+}]$$

$$=(E_{\mathbf{k}^+}+E_{\mathbf{k}^-})\frac{Z\mu}{V^2}. \quad (19)$$

These quasiparticles thus form a pseudogap where spin and charge matrix elements *vanish* on the Fermi surface and grow *linearly* with energy. In the special particle-hole symmetric case ($\mu=0$), these coherence factors vanish *throughout* the gap, forming a neutral band of excitations that only conduct heat. Since the paramagnetic spin and charge response functions of the quasiparticle fluid are proportional to the square of these matrix elements, the corresponding local response functions grow quadratically with energy

$$\frac{\chi''_{\mathrm{sp,ch}}(\omega)}{\omega}\propto\left[\frac{\omega}{T_K}\right]^2\left[\frac{\mu}{D}\right]^2. \quad (20)$$

This unusual energy dependence of matrix elements permits this state to mimic one with constant coherence factors, but a *linear* density of states (line of gap zeros).

We briefly list the main consequence of these results: (i) A large quasiparticle thermal conductivity in the absence of a quasiparticle contribution to the thermopower and electrical conductivity. (ii) Linear specific heat coefficient of magnitude $\gamma=\frac{1}{4}\gamma_n(1+\mu^2/V^2)$ where γ_n is the linear specific heat in the absence of the local moments. As μ varies γ can vary between values characteristic of a conventional metal, and values characteristic of a heavy fermion metal. (iii) A T^3 component to the NMR relaxation rate superimposed upon an activated background;

$$\frac{1}{T_1T}\propto\left[1+\frac{V^2}{T_K^2}\right]e^{-(T_K/T)}+\frac{\pi^2}{6}\left[\frac{T^2\mu^2}{V^4}\right]. \quad (21)$$

Since the spin matrix elements $\langle\epsilon|S^\pm|\epsilon\rangle=0$, the T^3 response is anisotropic and vanishes when the applied field is parallel to the $\hat{\mathbf{b}}$ axis.

Finally, we should like to mention the collective properties of this state. Some past studies of odd-frequency pairing have encountered a negative phase stiffness [18]. In our mean field theory, the phase has "coiled up" into a staggered configuration: This stabilizes the state and develops a positive phase stiffness. To compute the London response to a vector potential $\mathbf{A}$, we replace $\epsilon_{\mathbf{k}}\to\epsilon_{\mathbf{k}-e\mathbf{A}\tau_3}$. The London kernel $\Lambda\delta_{ab}=\nabla^2_{A_aA_b}F[\mathbf{A}]$ is then

$$\Lambda=-\frac{e^2v_F^2T}{6}\sum_\kappa\mathrm{Tr}[\mathcal{G}_{c\uparrow}(\kappa)^2-\mathcal{G}_{c\uparrow}(\kappa)\tau_3\mathcal{G}_{c\uparrow}(\kappa)\tau_3], \quad (22)$$

where the minus sign is a result of the staggered phase. Carrying out the energy integral at $T=0$

$$\Lambda=\frac{Ne^2}{4m}\int_0^{T_K}d\omega\frac{\Delta(\omega)^2}{[\Delta(\omega)^2+\mu^2]^{3/2}}, \quad (23)$$

where we have set $N/2m\equiv\frac{1}{3}\rho v_F^2$. In the special case of $\mu=0$, this integral simplifies to $\Lambda=(Ne^2/m)(T_K/4D)$. This small phase stiffness is consistent with the large coherence lengths $\lambda_L^{-2}=\mu_0\Lambda$ of heavy fermion superconductors, and may be regarded as a consequence of a condensation of "heavy electrons" with mass $m^*=mD/T_K$.

Macroscopic properties of the paired state are governed by slow rotations of the order parameter. In the absence of anisotropy, the long-wavelength action is spin rotationally invariant, given by a $\mathrm{U(1)}_{\mathrm{charge}}\times\mathrm{SU(2)}_{\mathrm{spin}}$ sigma model. Despite the phase stiffness, it cannot support a supercurrent without anisotropy. Spin isotropy means that the vacuum is not topologically stable against the creation of vortex pairs of the same sign: Uniform gradients of the phase can continuously develop to cancel an externally applied vector potential by twisting the order parameter "into the third dimension" [19]. The phase stiffness does guarantee a Meissner phase where fluxoids cannot penetrate, but an absence of topological stability means there is no rigidity to the flux phase. Anisotropy will stabilize the flux phase by aligning the order parameter with the crystal axes, lowering the symmetry to a $\mathrm{U(1)}_{\mathrm{charge}}$ X-Y model, where a macroscopic supercurrent is topologically stable.

Several questions of a technical nature surround our simple mean field theory. One performance benchmark of our mean field theory is provided by the one impurity Kondo model: Here we may compare its performance with the exact results and the well established large N mean field theory (MFT) [3]. For this model, the Majorana MFT correctly yields a unitary $\pi/2$ phase shift for the scattered electrons; it also predicts an enhanced isotropic susceptibility and linear specific heat: features consistent with the Fermi liquid fixed point. The mean field Wilson ratio $\chi/\gamma=8/3$ compares more favorably with the exact value $\chi/\gamma=2$ than the large N MFT, where $(\chi/\gamma)_{N\to\infty}=1$. As in the large N approach, RPA fluctuations of the phase variables develop power-law correlations in the mean field order parameter. Unlike the large N approach, power-law correlations are physically manifested as long-time correlations of the physical spin-pair operator $\mathbf{M}(t)=\mathbf{S}(t)\psi_\downarrow(t)\psi_\uparrow(t)\propto V(t)\times\sigma_2\boldsymbol{\sigma}V(t)$. Verification of power-law pair correlations $\langle\mathbf{M}(t)\mathbf{M}(0)\rangle\sim t^{-\alpha}$ $(\alpha\sim 1)$ would provide an independent test of incipient odd-frequency pairing in the Kondo impurity model [20]. Beyond the one impurity model, it remains to be seen whether our approach can also recover the normal phase by a careful treatment of these fluctuations.

Experimentally, the strong frequency dependence of coherence factors in our theory may help reconcile the observation of large linear specific heats and thermal conductivities in heavy fermions superconductors with the consistent *absence* of a corresponding Korringa NMR relaxation normally associated with gapless superconductivity.

In conclusion, we have examined an alternative treatment of the Kondo lattice model for heavy fermions that uses a Majorana representation of the spins. Our theory predicts a low temperature ground state with odd-frequency triplet pairing and surfaces of gapless neutral excitations. Spin and charge coherence factors vanish on the Fermi surface, predicting an intrinsic thermal conductivity and linear specific heat that coexist with a T^3 NMR relaxation rate. Independently of these issues, it provides a first stable realization of Berezinskii's odd-frequency pairing.

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